Electron Localization in $La_{2-x}Sr_xCuO_4$ and the Role of Stripes

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The normal-state in-plane resistivity (ρ_{ab}) is measured in lightly Zn-doped $La_{2-x}Sr_xCuO_4$ (LSCO) crystals with $0.06 \le x \le 0.17$ to systematically study the localization behavior. It is found that the localization temperature, $T_{\rm loc}$, where $\rho_{ab}(T)$ turns from metallic ($d\rho_{ab}/dT > 0$) to insulating ($d\rho_{ab}/dT < 0$), shows an anomalous enhancement at $x = 0.12$. Intriguingly, the doping dependence of $T_{\rm loc}$ in Zn-doped LSCO is found to be similar to that of Zn-free LSCO where superconductivity is suppressed by a high magnetic field. This suggests that the mechanism of the localization is the same in lightly Zn-doped LSCO and in Zn-free LSCO under magnetic field, and in both cases it is probably caused by the freezing of the electrons into an inhomogeneous state, which leads to the spin stripes at low temperatures.

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In underdoped $La_{2-x}Sr_xCuO_4$ (LSCO) superconductors, it is known that the temperature dependence of the in-plane resistivity $\rho_{ab}(T)$ becomes insulating at low temperatures when the superconductivity is suppressed by a high magnetic field,[1] and that the metal-to-insulator crossover in the normal state takes place at optimum doping.[2] Since it is unusual to have an insulating normal state in a superconductor, it would be of importance to understand the origin of this peculiar localization in underdoped LSCO. On the other hand, it is also known that a metallic transport ($d\rho_{ab}/dT > 0$) is realized in LSCO with only 1%-Sr doping at moderate temperatures.[3] These two peculiarities make a systematic study of the localization behavior, the transition from high temperature metallic ρ_{ab} to low temperature insulating ρ_{ab}, particularly meaningful in extending our knowledge on the electronic state of underdoped LSCO. Since the localization behavior was studied by the measurements in pulsed high magnetic field only for a few x values,[2] it is the purpose of the present work to study the x dependence in more detail to understand the mechanism of the localization.

Another peculiar feature in underdoped LSCO is the static spin stripes which develop at low temperatures and are observed by elastic neutron scattering experiments.[4,5] However, there is still no direct evidence for the presence of charge stripes in LSCO, although experimental results that indirectly indicate some charge inhomogeneity are accumulating.[6,7,8,9] Theoretically, various forms of charge inhomogeneity (or charge order) have been proposed to exist in cuprates.[10,11,12,13,14,15,16,17,18] If the charge inhomogeneity indeed exists in underdoped LSCO, the localization phenomena might be associated with a change in the charge dynamics due to the formation of such a structure, as is the case with the Nd-doped LSCO where static charge stripes are known to be stabilized and localization takes place below the temperature of low-temperature-orthorhombic (LTO) to the low-temperature-tetragonal (LTT) structural phase transition.[19,20,21]

In this work, we systematically study the localization behavior of underdoped LSCO using high quality single crystals where superconductivity is suppressed by Zn substitution. The amount of Zn doped into the Cu site is 2%, to avoid adversely affecting the pristine electronic state of LSCO. In fact, it was observed by μSR measurements that a larger amount of Zn substitution causes a significant suppression in the magnetic correlations.[22] We find that the localization temperature, $T_{\rm loc}$, where $d\rho_{ab}/dT$ turns its sign from positive to negative with decreasing temperature, shows an anomalous enhancement at $x = 0.12$, which is close to 1/8. Since the stripe ordering of spins is known to be pronounced at the 1/8 carrier concentration[4] and this order is expected to be triggered by some form of charge inhomogeneity,[10,11,12,13,14,15,16,17,18] the present result strongly suggests that the localization is caused by the freezing of the electrons into an inhomogeneous state that triggers the spin stripes at low temperatures.

TABLE I: Results of the ICP-AES analyses for grown $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ single crystals. $y = 0.02$ for each sample in nominal composition.

nominal x	0.06	0.08	0.10	0.12	0.15	0.17
actual x	0.059	0.078	0.097	0.117	0.145	0.165
actual y	0.014	0.015	0.018	0.016	0.017	0.017

The series of $La_{2-x}Sr_xCu_{0.98}Zn_{0.02}O_4$ single crystals (x = 0.06, 0.08, 0.10, 0.12, 0.15, and 0.17 in nominal composition) are grown by the traveling-solvent floating-zone technique.[23] To compensate Cu evaporation during the growth, we prepare raw rods with the cation ratio of La : Sr : Cu : Zn = $(2-x)$: x : 1 : 0.02 for each x. Inductively-coupled plasma atomic-emission spectroscopy (ICP-AES) is performed to determine the cation ratio in the grown crystals. Results are summarized in Table I. The measured x values are very close to the nominal ones, and thus in this paper we label the samples with nominal compositions for clarity.

The crystals are cut into rectangular platelets with

each edge parallel to the crystallographic axis within an error of 1° using x-ray Laue technique. Typical sample size is 2.0 × 0.5 × 0.15 mm^3, where the c-axis is perpendicular to the platelets. The samples are annealed at 800°C for 40 hours in air, followed by rapid quenching to room temperature, to remove oxygen defects in the as-grown crystals. The temperature dependence of ρ_{ab} is measured using a standard ac four-probe method under dc magnetic field parallel to the c-axis up to 18 T in the temperature range from 3.2 K to 300 K.

Figure 1 shows the temperature dependence of ρ_{ab} in zero and 18-T fields with the vertical axis in logarithmic scale. In zero field, except for the $x = 0.12$ sample (on which we elaborate later), the behavior of $\rho_{ab}(T)$ at moderate temperatures does not qualitatively change with x, which is the same as the behavior of Zn-free LSCO crystals.[3] In samples with $x \leq 0.10$, superconductivity is suppressed completely with 2%-Zn substitution and $\rho_{ab}(T)$ becomes insulating at low temperatures, which is also similar to the behavior of Zn-free underdoped LSCO when superconductivity is suppressed by pulsed high magnetic field.[1]

In order to additionally suppress superconductivity, 18-T magnetic field is applied along the c-axis. In samples with $x \leq 0.10$, no significant change is observed upon applying 18-T magnetic field. In the $x = 0.12$ sample, superconductivity is almost completely suppressed under the 18-T magnetic field and $\rho_{ab}(T)$ shows an insulating behavior down to low temperatures. In samples with

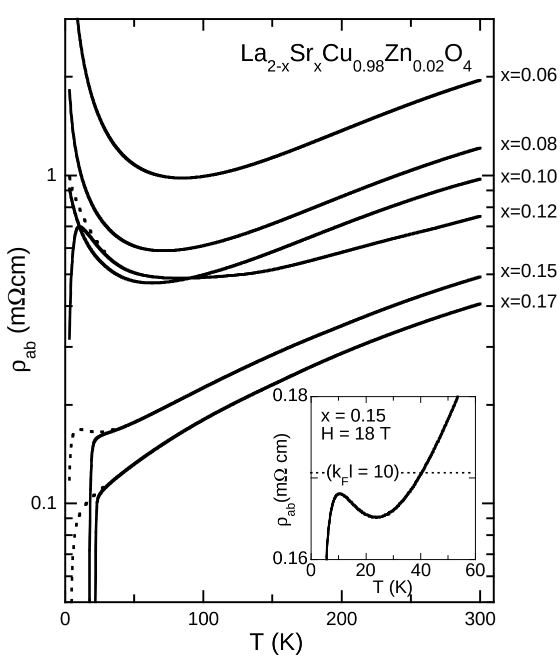

FIG. 1: Temperature dependences of ρ_{ab} for 2%-Zn-doped LSCO single crystals with $x = 0.06 - 0.17$. Solid lines and dotted lines represent the data in zero field and in 18-T field, respectively. Inset: Enlarged view of the data for the $x = 0.15$ sample in 18-T field.

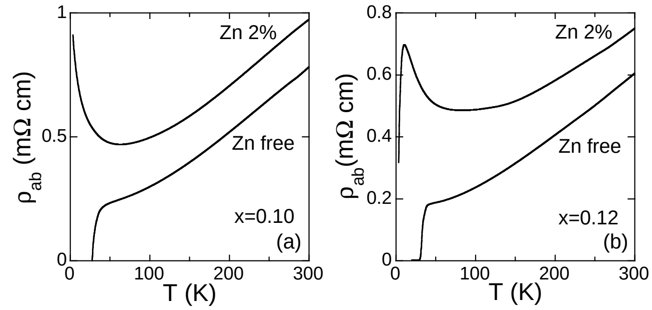

FIG. 2: Comparisons of $\rho_{ab}(T)$ between Zn-free and Zn-doped LSCO crystals at two representative dopings.

$x \geq 0.15$, superconductivity still shows up under 18 T, but we can observe a resistivity upturn below 25 K in the $x = 0.15$ sample. As is shown in the inset to Fig. 1, this resistivity upturn takes place in quite a clean system where $\rho_{ab} \simeq 170$ $\mu\Omega$cm. If we convert this resistivity into $k_F\ell$ (k_F is the Fermi wave number and ℓ is the mean free path) by using the relation $k_F\ell = hc_0/\rho_{ab}e^2$ (c_0 is the interlayer distance), which is derived by assuming the Luttinger theorem and the Drude-type transport in two-dimension, we obtain $k_F\ell \simeq 10$. This value of $k_F\ell$ is unusually large for a system with localization behavior.[2] On the other hand, our previous measurements showed that $\rho_{ab}(T)$ exhibits unusually metallic behavior at moderate temperatures in slightly hole-doped LSCO samples where $k_F\ell \simeq 0.1$.[3] Given that a metal (insulator) would normally be expected for $k_F\ell \gtrsim 1$ ($k_F\ell \lesssim 1$), these peculiar transport properties suggest that the electronic structure in LSCO cannot be described within the conventional band picture; we have therefore argued[3] that a self-organized charge inhomogeneity might be responsible for this puzzle. Note that this unusual localization behavior is observed not only in LSCO but also in $YBa_2Cu_3O_y$ system[24] and in $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ system.[25]

Figure 2 shows a comparison of $\rho_{ab}(T)$ in zero field for Zn-free and Zn-doped samples at $x = 0.10$ and 0.12. The data for Zn-free samples are from our previous results.[3,23] In the normal state, Zn-substitution usually causes temperature-independent additional scatterings in the in-plane transport in cuprates,[24,26] which is observed as a parallel shifting of $\rho_{ab}(T)$ in the $x = 0.10$ sample in Fig. 2(a). However, as is shown in Fig. 2(b), in the $x = 0.12$ sample the slope of $\rho_{ab}(T)$ changes in the Zn-doped sample. It seems as if a resistivity of different origin grows below ~150 K in the $x = 0.12$ sample. This resistivity behavior is rather similar to that of Zn-free LSCO where superconductivity is suppressed by high magnetic field;[2] namely, at $x = 0.12$, an unusually large magnetoresistance has been observed below ~100 K under 60-T magnetic field.

To map out the localization behavior in the phase diagram, we define the temperature where $\rho_{ab}(T)$ shows a minimum value as the localization temperature $T_{\rm loc}$. Figure 3 shows the x dependences of $T_{\rm loc}$ for 2%-Zn-doped

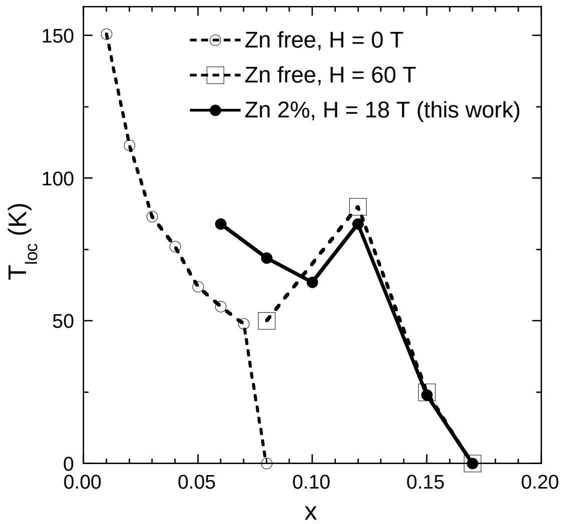

FIG. 3: x dependences of $T_{\rm loc}$ for Zn-free and 2%-Zn-doped LSCO single crystals. Open circles and squares denote $T_{\rm loc}$ of Zn-free LSCO.[2,3,23,27] Solid circles represent $T_{\rm loc}$ of Zn-doped LSCO (this work); note that the 18-T field affects $T_{\rm loc}$ only in the $x = 0.15$ samples.

LSCO crystals together with the data obtained from the previous results for Zn-free LSCO crystals.[2,3,23,27] The overall tendency of $T_{\rm loc}$ in Zn-doped samples is quite similar to that in Zn-free samples: $T_{\rm loc}$ tends to decrease with increasing x and disappear at $x = 0.17$, and it is clearly seen that $T_{\rm loc}$ is anomalously enhanced at $x = 0.12$ in both Zn-doped and Zn-free samples. At this carrier concentration, the temperature where the static spin stripes are formed is also enhanced, which was observed by elastic neutron scattering experiments,[4] and thus the enhancement of $T_{\rm loc}$ at $x = 0.12$ indicates that the localization observed in resistivity has relevance to the spin stripes. In recent μSR measurements,[28,29] the temperature where the Cu spin dynamics changes, which we call T_0 here, was analyzed in a series of LSCO samples and this T_0 was found to show a doping dependence similar to that of $T_{\rm loc}$. This fact further supports the idea that the localization is related to the formation of the spin stripes, which is reflected in the Cu spin dynamics. In passing, the similarity of the role of Zn doping to that of high magnetic fields demonstrated in Fig. 3 motivates one to estimate the field equivalence of the 2%-Zn doping: comparison of the present data to those of Refs. 1,2 suggests that the 2%-Zn doping roughly corresponds to 40 T.[30]

There are other results which we consider to support the relation between the localization and the spin texturing. Lake *et al.*[31] observed an enhancement of the incommensurate magnetic peaks in underdoped LSCO by magnetic field in their elastic neutron scattering experiments. Correspondingly, Sun *et al.*[32] and Hawthorn *et al.*[33] observed decreasing quasiparticle thermal conductivity with increasing magnetic field in underdoped LSCO crystals at very low temperatures. Taken together, these results indicate that a magnetic-field-induced localization of quasiparticles[32] is associated with a magnetic-field-induced spin order, and this quasiparticle localization in the superconducting state is probably responsible for the insulating behavior under high magnetic field.

It is useful to note that the antiferromagnetism is competing with superconductivity in cuprates,[12,14] and both the magnetic field[31] and the Zn doping[34] enhance the antiferromagnetism; in this sense, the similarity between the roles of the two is rather natural. Also, it is widely conjectured[10,11,12,13,14,15,16,17,18] that the magnetic state that is competing with superconductivity involves some texturing of spins[31] and charges.[35,36] Our data point to an intimate link between the spin stripes and the charge localization, which can be naturally understood if some charge inhomogeneity triggers both the spin stripes and the localization. Intriguingly, a charge-inhomogeneous state has certain similarities to a granular system, for which the $\log(1/T)$ resistivity has been experimentally observed[37] and theoretically proposed[38]. Thus, we speculate that the peculiar localization in underdoped cuprates may originate from an effective granularity brought about by some form of charge inhomogeneity when the superconductivity is suppressed.

In passing, one can see in Fig. 3 that $T_{\rm loc}$ of 2%-Zn-doped LSCO with $x > 0.10$ is almost identical to that of Zn-free LSCO under high magnetic field, while for $x < 0.10$, $T_{\rm loc}$ of Zn-doped samples is about 20 K higher than that of Zn-free ones. This difference would be related to the Fermi-surface topology elucidated by the angle-resolved photoemission spectroscopy (ARPES) measurements:[39] In slightly Sr-doped LSCO samples, only the Fermi arc is observed at the Fermi energy E_F near the $(\pi/2,\ \pi/2)$ position in the Brillouin zone, and electronic states near $(\pi,\ 0)$ are located deeply below E_F,[40] which suggests that the electronic transport is governed by the quasiparticles on the Fermi arcs in slightly hole-doped LSCO crystals.[41] As x increases, the $(\pi,\ 0)$ band moves up and touches E_F at $x \sim 0.10$,[40] above which the electrons near $(\pi,\ 0)$ will participate in the transport. Therefore, if the Zn-induced scatterings of the quasiparticles on the Fermi arcs are stronger than those on the $(\pi,\ 0)$ band, the localization effect in $x < 0.10$ samples, where the quasiparticles are confined on the Fermi arcs, would be more pronounced. Further experiments would be desirable to clarify the details of the possible scattering-rate anisotropy.

Finally, let us discuss the temperature dependence of the insulating behavior. Figure 4 shows $\log(1/T)$ plots of $\rho_{ab}(T)$ of Zn-doped LSCO with $x \leq 0.12$. One can see the $\log(1/T)$ insulating behavior in the $x = 0.12$ sample (where the 18-T field seems insufficient to suppress superconductivity completely below 10 K), while for $x \leq 0.10$ the resistivity diverges faster than $\log(1/T)$. On the other hand, the transport study under 60-T field showed that in Zn-free LSCO at $x = 0.08$, the resistivity exhibits clear $\log(1/T)$ divergence.[1] Therefore, for the

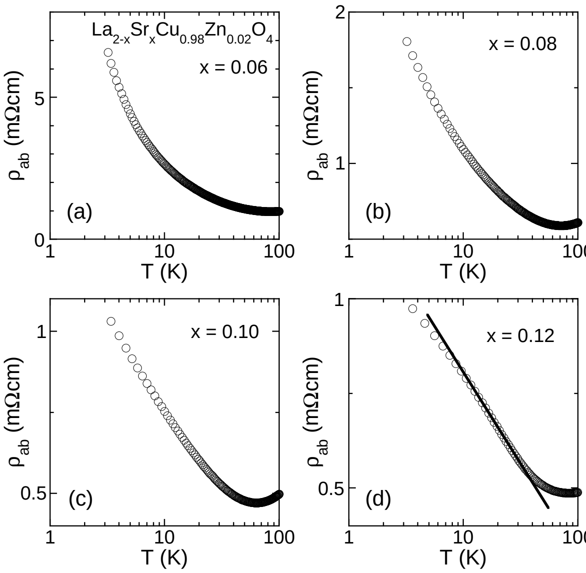

FIG. 4: log(T) plots of $\rho_{ab}(T)$ for Zn-doped LSCO with $x \leq 0.12$.

same Sr content, the insulating behavior is slightly different in the two cases. It is useful to note that in strongly insulating samples where ρ_{ab} exceeds a rough criterion of 2–3 mΩcm (which corresponds to $k_F\ell \sim 1$), $\rho_{ab}(T)$ becomes roughly consistent with the variable range hopping (VRH) behavior; actually, the data for $x = 0.06$ [Fig. 4(a)] is consistent with the VRH behavior. On the other hand, the $\log(1/T)$ behavior is observed whenever the resistivity is less than the criterion in Zn-free underdoped samples. The 2%-Zn-doped samples at $x = 0.08$ and 0.10 [Figs. 4(b) and 4(c)] are peculiar in that their $\rho_{ab}(T)$ behavior is neither $\log(1/T)$ nor VRH, even though their resistivity is smaller than the criterion. If the $\log(1/T)$ behavior is associated with an effective granularity as we conjectured above, the slight anomaly in the Zn-doped samples may indicate that the Zn impurities not only stabilize the spin/charge texturing but also disturb the local electronic states.

In summary, the insulating behavior of underdoped LSCO is studied in a series of single crystals where superconductivity is suppressed by 2%-Zn doping. It is found that the localization temperature, $T_{\rm loc}$, is anomalously enhanced at $x = 0.12$. Intriguingly, the x dependence of $T_{\rm loc}$ is found to be similar to that of Zn-free LSCO under high magnetic fields. This suggests that the localization mechanism is essentially the same in Zn-doped LSCO and in Zn-free LSCO under magnetic field. Furthermore, the behavior of $T_{\rm loc}$ is rather similar to that of the spin freezing temperature determined by μSR measurements. Based on these results, we discuss possible relevance of the spin/charge texturing to the localization phenomena.

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